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Urban scale modeling of particle number concentration in Stockholm

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Abstract

A three-dimensional dispersion model has been implemented over the urban area of Stockholm (35×35 km) to assess the spatial distribution of number concentrations of particles in the diameter range 3–400 nm. Typical number concentrations in the urban background of Stockholm is 10 000 cm^{-3}, while they are three times higher close to a major highway outside the city and seven times higher within a densely trafficked street canyon site in the city center. The model, which includes an aerosol module for calculating the particle number losses due to coagulation and dry deposition, has been run for a 10-day period. Model results compare well with measured data, both in levels and in temporal variability. Coagulation was found to be of little importance in terms of time averaged concentrations, contributing to losses of only a few percent as compared to inert particles, while dry deposition yield particle number losses of up to 25% in certain locations. Episodic losses of up to 10% due to coagulation and 50% due to deposition, are found some kilometers downwind of major roads, rising in connection with low wind speed and suppressed turbulent mixing. Removal due to coagulation and deposition will thus be more significant for the simulation of extreme particle number concentrations during peak episodes.

The study shows that dispersion models with proper aerosol dynamics included may be used to assess particle number concentrations in Stockholm, where ultrafine particles principally originate from traffic emissions. Emission factors may be determined from roadside measurements, but ambient temperature must be considered, as it has a strong influence on particle number emissions from vehicles.

Keywords: Coagulation; Dry deposition; Dispersion model; Aerosol model; Traffic; Emission factor; Particle number concentrations; Particle size distribution

1. Introduction

Based on established links between monitored particle mass concentrations and health effects, EU member countries are now obliged to comply with specified maximum levels of PM10 (mass of particles with diameter $<10\,\mu m$). Recent evaluations of the World Health Organization (WHO, 2003) suggest that some health effects show stronger links to the fine fraction and recommend the regulation of PM2.5 instead of PM10. WHO also states that more research is needed on possible links between health and other metrics that describe airborne particles, such as total number concentration (ToN). The latter is more dominated by local combustion sources than PM2.5, implying stronger

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concentration gradients over an urban area. While annual mean PM2.5 levels are almost the same in the rural and urban background, and only show slightly increased levels at curb-side locations (Areskoug et al., 2001), particle number concentrations are several times higher in the urban background as compared to the rural background and more than 10 times higher at curb-side (Ketzel et al., 2003b). Urban background here means the concentration found inside the city, but at roof level or at a ground level location distant from major roads or other local sources.

Total number concentrations in diesel exhausts are dominated (>90%) by nanoparticles smaller than 50 nm in diameter, composed of metals, elemental carbon and semi-volatiles formed during dilution and cooling after leaving the exhaust pipe (Kittelson et al., 2000). Recent findings indicate that as much as as 95% of the volatile component of diesel particles consists of unburned lubrication oil (Sakurai et al., 2003). Spark ignition combustion yields even smaller particles than diesel (Ristovski et al., 1998) and gasoline vehicles furthermore emit at a highly variable rate that depends on engine load and cruising speed (Maricq et al., 1999; Andersson and Wedekind, 2001).

The fate of ultrafine particles, i.e. those with diameter <100 nm, over the urban scale has been discussed from a theoretical point of view by Zhang and Wexler (2002). Relevant processes that may affect total number concentrations are condensation/evaporation, coagulation and deposition, the latter either through dry deposition to the ground or building surfaces or through scavenging by rain. Condensation/evaporation does not change total particle number, but it changes the size of the particles, which is important for the coagulation and deposition rates. According to the time scale analysis for urban aerosols by Zhang and Wexler (2002), condensation will be the leading process for particles larger than 50 nm and below 2 μm, while coagulation will only affect particles <50 nm.

Wehner et al. (2002) used a sectional aerosol model to interpret shifts in peak size distributions from about 15 nm at curb-side to 20–25 nm in an urban background station, suggesting condensation and coagulation to be relevant processes over the urban scale. Ketzel et al. (2003b) interpreted a similar shift, from 22 nm at curb-side to about 28 nm in the urban background, together with a reduced particle number to NO_x ratio (assuming NO_x to be inert on the urban scale), as a result of deposition, coagulation and condensational growth. They also gave an estimate of the particle removal to be on the order of 15–30%, going from curb-side to an urban background station.

The urban model study to be presented here is based on experiences from three earlier studies of Stockholm traffic environments, in a car tunnel (Gidhagen et al., 2003, hereafter G1), a street canyon (Gidhagen et al., 2004a, hereafter G2) and close to a highway (Gidhagen et al., 2004b, hereafter G3). The three studies all indicate that on the local (street) scale, coagulation is only effective at conditions of very high concentrations, such as those found in the car tunnel or very close to the vehicle exhaust pipe. When the vehicle exhaust particles reach an open air monitor, situated, e.g. on the curb-side of a street canyon, the dilution has already reduced the concentrations so much that coagulation is slow. Dry deposition was shown to be most effective very close to the vehicles, where the vehicle movements contribute to high friction velocities and turbulence that increase the deposition efficiency. In the street canyon study G2 the removal due to the combined action of deposition and coagulation taking place between a curb-side monitor and a roof level monitor was estimated to be less than 10% (G2). This is of little significance when the overall uncertainties in ToN emission factors are taken into account. In open road conditions, like those close to the highway discussed in G3, the rapid dilution and lower background concentrations makes coagulation even less important. These results indicate that an urban model could use emission factors based on measured concentrations in open air and close to roads, calculated by inverse modeling and assuming inert particles. The removal processes taking place between the vehicle exhaust pipe and the nearest monitor, will in that case be incorporated in an effective emission factor that depend on vehicle composition, vehicle speed and ambient temperature. Table 1 summarizes a number of ToN emission factors determined from roadside measurements. Emission factors for total particle number based on laboratory measurements are strongly dependent on dilution rates (Kittelson et al., 1999), which make them difficult to use for dispersion modeling purposes.

Table 1 shows rather consistent factors for HDV (heavy duty vehicles), while the ranges are somewhat larger for LDV (light duty vehicles). The G1 study reported emission factors based on model simulations that included coagulation and deposition, while the other factors are all given assuming inert particles. The Stockholm G2 and G3 emission factors are well within the range of other values reported. The ambient temperature (Kittelson et al., 2000; Charron and Harrison, 2003) and engine load (Maricq et al., 1999) and the different lower cutoffs of the monitors are factors that must be taken into account in a comparison. As for the temperature effect, an analysis of the ToN/NO_x ratio in a Stockholm street canyon environment indicates—assuming NO_x factors to be temperature independent—that the ToN emission factor during a cold winter day may be up to twice as big as during a warm summer day (G2). Similar indications were found during short-term measurements close to a highway north of Stockholm (G3).